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A broad research program was carried out aimed at the study of advanced superconducting electronic materials, superconducting tunnel junctions and the tunneling process itself. An artificially deposited amorphous silicon tunnel barrier was developed and applied (1) to tunneling studies of the A15 class superconductors Nb_3Sn and V_3Ga, and (2) to studies of the role of localized states on the tunneling process. The first practical sandwich-type superconducting/normal/superconducting Josephson junction device was developed based on a Nb-alloyed amorphous Si barrier materials near a metal/insulator transition. The device appears to be an attractive alternative to shunted tunnel junctions for many applications. Finally, the first practical approach to the deposition of the new high-temperature oxide superconductors was developed—the so-called post-annealed approach—and the materials science of the resulting films studied in detail. Tunneling studies of the high-temperature oxide superconductors were also carried out and provided the first evidence for the very large superconductive energy gaps now commonly reported for these materials.

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ELECTRICAL TRANSPORT THROUGH TUNNEL BARRIERS AND THIN DIELECTRIC LAYERS AND PHYSICAL PROPERTIES OF THE HIGH Tc OXIDE SUPERCONDUCTORS

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I. Introduction and Summary of Research Completed

In broad terms the research supported under this contract was aimed at the study of advanced superconducting electronic materials, tunneling studies of these materials and studies of the tunneling process itself. It also involved the development and use of advance vapor deposition techniques to synthesize superconducting materials in thin film form and to create artificial (and artificially structured) barriers in superconducting tunnel junctions and other types of Josephson junctions.

Historically, there were four basic elements in the program. The first was the development of approaches and procedures for forming superconductive tunnel junctions on the (then) high transition temperature superconductors of the A15 class, and the study of the characteristics of these devices from both the practical and fundamental points of view. This work led to a general approach for forming tunnel junctions on materials with poor native oxides through the use of ultra-thin, deposited amorphous silicon (a-Si) layers that were subsequently oxidized. These deposited barriers were found to be widely applicable—a kind of generic tunnel barrier material—and opened up the field of tunneling studies of the A15 and many other superconductors by ourselves and other groups. As part of this program, we applied these barriers to tunneling studies of Nb_3Sn and V_3Ga—two important A15 superconductors. The work clarified the origins of the high transition temperatures of these two superconductors and, in particular, the importance of disorder in the reduction of Tc as their composition deviated from the ideal 3:1 stoichiometry.

The second element was a comprehensive study of electronic transport through tunnel barriers containing localized states. This work was an outgrowth of our development of a-Si tunnel barriers described above. Amorphous silicon is known to contain a high density of localized states. By using various forms of our deposited amorphous silicon barriers, we were able to

study transport via such localized states as a function of their density, of their location in the barrier, and of the thickness of the barrier. Ultimately we learned how to deposit pin-hole-free, pure a-Si barriers with no oxide. These various a-Si barriers turned out to be an effective model system for studying the effects of localized states on the tunneling process. The results have led to an increasingly thorough understanding of the role of localized states on tunneling, their relation to resonant tunneling and how they lead to a deterioration of superconductive tunneling device characteristics. The results of this work are also relevant to transport in mesoscopic systems (e.g., one-dimensional MOSFETS). It has stimulated a sizeable amount of theoretical work that has played a crucial role in interpreting the data.

The third element of our program was the study of transport through barriers formed from materials near a continuous insulator/metal transition. The material system employed was an amorphous Nb-Si alloy. This alloy undergoes an insulator/metal transition at about 11% Nb. The goal was to understand transport at short length scales in materials exhibiting electronic localization, and to investigate potential device applications. By incorporating barriers just on the metal side of the insulator/metal transition, we were able to form for the first time sandwich-type superconducting/normal/superconducting (SNS) Josephson devices with usefully high resistances (e.g. ohms). Previous sandwich-type SNS junctions were made using elemental normal metal barriers (e.g., copper or gold) and had exceedingly low resistances. Our high resistance SNS sandwich-type device is an attractive alternative to the traditional resistively shunted tunnel junctions for applications where nonhysteretic I-V characteristic are required. Our device is much more compact and more easily fabricated than the shunted tunnel junctions. It also may have better performance.

These studies of tunneling via localized states, and the nature of the transport in barriers near an insulator/metal transition, may be of relevance for recent work aimed at forming tunneling or Josephson junction devices with the new high temperature oxide superconductors. The most obvious examples are the attempts to use $PrBa_2Cu_3O_7$ as a barrier in sandwich-type $YBa_2Cu_3O_7$/ $PrBa_2Cu_3O_7$/$YBa_2Cu_3O_7$ tunnel junctions. Transport in the insulating phases of these oxide superconductors is thought to occur via variable range hopping, and

therefore localized states are implicated. We expect that this work may also have relevance for the conductivity and breakdown of thin insulating layers in integrated circuits.

The fourth element of this program was a response to the discovery of superconductivity at very high temperatures by Bednorz and Mueller. Since that astonishing event, we have included studies of these remarkable new superconductors in our work. It represents a natural extension of our previous program. In collaboration with our colleagues at Stanford (Geballe, Kapitulnik and Hammond), we developed the first successful approach to the deposition of these new superconductors in thin films form and have studied the properties of the resultant films from various points of view. Among the highlights of this work were the first synthesis of these materials in thin film form, development of the post-annealed process for film synthesis, the first detailed study of the materials science of post-annealed films, the first demonstration of the high current carrying capacity of these superconductors, and tunneling studies using point contacts and sandwich-type junctions that first suggested and continue to suggest anomalously large energy gaps in these superconductors. In collaboration with the groups of John Clarke and Paul Richards at the University of California at Berkeley, we have also studied the noise processes in these films and established the dependence of this noise on the microstructure of the films. Finally, more recently we have explored the potential of electron beam reactive evaporation for the *in situ* growth of $YBa_2Cu_3O_7$. A useful summary of this work, along with that of other workers in the field, can be found in the review article prepared under this program for inclusion in the recent Special Issue of the Proceedings of the IEEE on Superconductivity.

In the remainder of this report, we discuss in turn each of these accomplishments in greater detail. Detailed references are included for the reader seeking additional detail. A list of all publications prepared under this contract is included in Appendix A.

II. Tunnel Junctions and Tunneling into the A15 Superconductors.

Superconductive tunnel junctions are important in both the electronic applications of superconductivity and as a remarkably effective probe of the

microscopic aspects of superconductivity. Traditionally, tunnel junctions could only be made on those superconductors for which the native oxide formed a good tunnel barrier. Unfortunately, useful barriers do not form naturally on the surface of most currently interesting superconducting materials. As we had shown in a previous ONR program, oxidized, deposited amorphous silicon (a-Si) barriers can be used to form tunnel barriers on a wide variety of sensitive materials in which the barrier properties are largely independent of the base material [1]. The idea is that the oxidized silicon (SiO_x), which actually forms the barrier, is separated from the surface of the material by an unoxidized layer of amorphous silicon. This a-Si layer serves to protect the underlying metal.

In the work supported by this program, this oxidized a-Si barrier was systematically applied to tunneling studies of Nb_3Sn [2,3,4] and V_3Ga [4,5,6]. A second essential part of this work was the development of deposition procedures necessary to grow thin films of these superconductors suitable for tunneling studies and device applications.

In the case of Nb_3Sn, we were able using electron-beam coevaporation to fabricate and study tunnel junctions over a wide range of Nb and Sn concentrations. Thus it was possible to study how the superconducting parameters of this materials evolved as one approached the ideal 3:1 stoichiometric material as the Sn concentration was increased. In particular, it was possible to study how the electron-phonon spectral function $\alpha^2F(\omega)$, which can be derived from the tunneling I-V characteristic and which is the quantity that most clearly reveals the microscopic origins of the superconductivity, evolved as a function of composition. The thin film deposition procedures and junction fabrication techniques developed are described in detail in Refs. 2 and 4. The detailed study of the electron-phonon spectral function is described in Refs. 3 and 4.

Our essential result is that as the 3:1 stoichiometric composition is approached, the spectral weight of the electron-phonon interaction shifts to lower frequencies and increases overall in strength. The net result is an increase in the electron-phonon interaction parameter. At the same time, the Coulomb repulsion μ^* is constant. These results reveal in microscopic terms why Tc increases as stoichiometry is approached. At a deeper level they show that it is

changes in the electron-phonon interaction, and not the electron-electron Coulomb interactions, that governs the systematics of Tc, and that mode softening alone is not enough to account for the increase in Tc as stoichiometry is approached. There must also be an overall increase in the density of states.

The case of V_3Ga is even more illuminating, since the A15 phase exists on both sides of stoichiometry. By studying the variation of Tc and and the transport properties of the Va-Ga system as we varied the composition through the 3:1 stoichiometry, we found that the most natural variable seemed to be resistivity, not the composition. More specifically, we found that the depression of Tc in off-stoichiometric material was a universal function of the resistivity, independent of the composition. This result strongly suggests that the Tc reduction is largely due to disorder and only secondarily to changes in electron density. Combining these observations with those we obtained on Nb_3Sn, we conclude that the reduction of Tc in off-stoichiometric A15 superconductors must be due primarily to the reduction of the peak in the electronic density of states near the fermi level brought on as a result of disorder. We also examined the electron-phonon spectral function for this material in order to establish whether phonon softening as observed in Nb_3Sn was also present in V_3Ga. These results are described in Refs. 5, 6 and 7.

Finally, in the case of V_3Ga we were able observe a splitting of the superconducting tunneling density of states in the presence of a high magnetic field—essentially a Zeeman splitting of the quasiparticle states in the superconductor. This represented the first observation of this effect in an A15 superconductor and confirmed that the Cooper pairs are in the usual singlet spin state. The effect would be absent in the case of triplet pairing and is smeared out in materials with strong spin-orbit scattering. V_3Ga was a particularly attractive material for this study because it is formed from low-Z elements and therefore has little spin-orbit scattering. It would be of considerable interest to repeat this experiment on the high Tc oxide superconductors, where the relevant constituent elements (Cu and O) have even lower Z's. These results are described in Refs. 6 and 7.

III. Transport Via Localized States in Tunnel Barriers

Transport via localized states in tunnel barriers is an oft quoted source of various non-idealities in superconductive tunnel junctions, e.g., excess conductance below the superconducting energy gap and reduction of the Josephson tunneling current. Since no good understanding of the effects of such processes was available, localized states became a "rug" under which many unexplained tunneling anomalies have become swept.

In a series of studies using variants of our a-Si barrier, various barrier configurations (a-Si/SiOx bilayers, SiOx/a-Si/SiOx trilayers, SiOx/a-Si:H/SiOx trilayers and pure a-Si monolayers) were produced and used to separate the direct tunneling processes from those involving localized states and to establish conditions in which transport via the localized states dominate the total tunneling current [7,8,9]. Also, by making micron-sized junctions, we were able to observe the contributions of individual localized states in the overall I-V curve [9]. With such small junctions it was possible to measure the energy dependence of the transmission via an individual localized state. We also studied the 1/f resistance fluctuations in these junctions associated with the localized states [10].

Among our principal observations were that:

(1) as expected theoretically, localized states contribute most when they are located at the electrical center of a barrier (i.e., when they are located where the tunneling probability to the two metal electrodes is equal) [7,9];

(2) as also expected theoretically, the dominant tunneling mechanism crosses over from direct tunneling to tunneling via localized states as the thickness of the tunnel barrier increases [7,8,9];

(3) in submicron junctions it is possible to isolate the conductance due to individual localized states, and the energy width of the transmission of these individual localized states is wider than that expected on the basis of the simplest theories of elastic (energy conserving) resonant tunneling. The width is also

temperature dependent, which suggests that inelastic processes are operative [7,9];

(4) tunnel barriers containing localized state exhibit very nonlinear I-V characteristics. This nonlinearity grows as the junctions are made thicker and the relative contribution of tunneling via the localized state increases. Previous workers attributed this nonlinearity to a low barrier height in a-Si (as small as 30 meV) [7,11]. Our results show that this interpretation is not correct and that the nonlinearity is due to the transport process via the localized states themselves [12]. Recently a theory of this nonlinearity, motivated by our observations, was presented that accounts for this nonlinearity in terms of sequential hopping via increasingly long chains of localized states as the bias voltage increases [13]. This process is the precursor of the variable range hopping process that governs electrical transport in bulk a-Si. Elaborations of the theory also predict a host of other phenomena that should be observable with our a-Si barriers. These include correlated tunneling process due to coulomb interactions that prevent multiple occupancy of the localized states during the tunneling process, and a Kondo resonance-related anomaly near zero bias. The latter effect results from an interaction of the conduction electrons in the electrodes with the spin of the electrons occupying the localized states.

(5) transport via localized state appears to be suppressed for now bias voltages over a voltage range that depends on the barrier thickness. There is no clear interpretation of this effect at the present time [11].

In addition to the results enumerated above, we also carried out studies of how the superconductive tunneling characteristic of Nb/a-Si/Nb tunnel junctions deteriorated as the contribution of tunneling via localized states was increased. The existence of these results has also stimulated recent theoretical work [14]. Most interesting are predictions of enhanced Josephson currents, nonsinusoidal current phase relations and even a prediction that $I = - I_0 \sin \phi$ under certain conditions!

From the above it should be clear that the physics of tunneling via localized state is richer than even we imagined when we began this program.

IV. Barriers Formed from Materials Near an Insulator/Metal Transition.

Transport in materials near a continuous insulator/metal transition has been the subject of intense interest. It is known that electrons become localized in such systems due simply to disorder. The role of Coulomb interactions introduces new elements to the problem that are much less well understood. In our program, we have focused on the behavior at short length scales where the conductance is length dependent.

The material system we chose was an amorphous Nb-Si alloy, because of the hope that in the long run we could make connection with our work on pure a-Si described above. Using deposition procedures similar to those we used for our very thin a-Si barriers, Nb/a-(Nb-Si)/Nb trilayers were formed and then patterned into junctions a few microns in diameter. Because of the very high resistances that we could achieve with these systems, we chose early in the program to explore the potential of our structures as high-resistance, sandwich-type Josephson junctions. A brief description of these devices and their electrical performance is contained in Ref. 15. A more complete description can be found in the thesis of Adrian Barrera, a visiting graduate student from the University of Mexico, who carried out the work [16]. These junctions readily yielded devices with resistances in the range of one ohm and I_cR products (a critical device figure of merit) of about 100 μV. The critical currents of these devices exhibited classical, single-slit-like magnetic diffraction patterns. They also exhibited cavity-induced rf Josephson steps in their I-V characteristics at voltages corresponding to 100 GHz radiation. Thus they are capable of very high- frequency application. In addition to these device studies, the various basic physical parameters of the barrier were also measured. These included the magnitude and temperature dependence of the proximity effect coherence length of the a-(Nb-Si).

As stated in the introduction, these devices represent an important expansion of the types of Josephson devices that are now available for applications. They are most attractive in those applications where shunted tunnel junctions are currently used. In this case, an external shunt is used to eliminate the hysteresis normally present in the I-V curve of a tunneling Josephson junction. The price paid is the space required to form the shunt and the markedly reduced I_cR-product of the device. Our junctions have comparable

or better I_cR-products, even in their unoptimized form, and are very compact. They appear ideal for use in arrays of Josephson junctions of interest for local oscillator or variable series-connected JJ delay line applications. In the case of local oscillators, it is desirable to have all the junctions contained within one wavelength, and for variable delay transmission lines, several junctions per wavelength in order to avoid distributed effects in the operation of such devices.

Finally we note that our approach may be applicable to the new high-Tc oxide superconductors, with which tunneling junctions may not be possible.

V. Deposition of the New Oxide Superconductors in Thin Film Form

Right after the confirmation of high temperature superconductivity in $(La\text{-}Sr)_2CuO_4$ by the University of Tokyo group, we immediately began attempts to grow these new superconductors in thin film form. These materials present problems of film growth quite unlike those of conventional superconductors. First of all, they contain up to five metallic elements, plus oxygen. Second, being oxides, they require deposition in considerable background pressures of oxygen, which presents many new technical problems. To get around these problems, in our original approach we simply deposited the metallic elements in a sufficient background pressure of oxygen to form chemically-stable amorphous mixtures of the various oxides of the metals and then post-annealed the films in oxygen at high temperature to form the desired high temperature Perovskite oxide superconducting phase. If suitable substrates are used (e.g., $SrTiO_3$), the resultant film growth is in the form of polycrystalline epitaxial growth.

It was with this so-called post-annealed approach that we deposited the first thin films of these new superconductors. We then went on to more systematically study the materials science of these films, the utility of various substrates, and the effects of substrate temperature on the properties of the resultant films. A detailed account of this work can be found in Ref. 17. The post-annealed process has been extended to many of the other high-temperature oxide superconductors by other researchers and is now one of the standard approaches.

It became clear early on, however, that it would be very desirable to grow the materials directly at low temperatures and on a wider range of substrates. This has led to various attempts to grow films of these materials *in situ*. A nearly uncountable number of papers have now been published on this subject, most of which report the empirical results of deposition by this or that technique and under various conditions. In our work, we have focused on electron-beam reactive coevaporation, and on systematic studies aimed at establishing quantitatively the conditions necessary for successful film growth. In particular, we have explored the relative utility of various forms of reactive oxygen, including atomic oxygen, charged ions and ozone, in addition to molecular oxygen [18].

As a part of this work, we have developed various means of introducing these reactive oxygen species into our evaporation system and to measure quantitatively their fluxes at the substrate. At the same time, the presence of a high background pressure of oxygen has necessitated the development of new approaches to monitoring the rates of the metal fluxes. Most of the conventional approaches cannot be used in such high background pressures (e.g., ion gauge rate monitors) or when the metallic species in question is oxidized on the detector surface (e.g., crystal quartz monitors). To solve these problems, in collaboration with Xinix, Inc, we have been exploring the use of atomic absorption rate monitoring [19]. This approach has the distinct advantage of being species selective and therefore to first-order independent of the high oxygen background pressure. The main problem we have encountered in utilizing this technique is the construction of thermally-stable mechanical fixtures (in the presence of substrate heaters) needed to hold the optical fibers and collimate the transmit light used to monitor the evaporant flux.

VI. Physical Properties of Thin Films of the High-Temperature Superconductors.

Using the thin films described above, and some bulk crystals produced by other member of our group under other contracts, under this program we have carried out various studies of the physical properties of these new superconductors. One major theme has been tunneling studies of these materials. We have used point-contact tunneling, scanning-tunneling

microscopes (STM), and conventional sandwich-type tunnel junctions. Some of the major findings of this work are:

(1) evidence for large energy gaps in these materials corresponding to values of $2\Delta/kTc = 6\text{-}7$. We were the first to report such large values, and subsequently many workers have reported similar results [20,21,22,23,24]. See Ref. 24 for a useful review of these results

(2) observation of anomalous conductance in the normal state tunneling in these materials at bias voltages well above the suspected energy gap region and for temperatures above Tc [See Ref. 24].

(3) STM images of the surface of 2212 BiSrCaCuO that showed the 27.2 Å superstructure present in these materials. This work also provided the first direct evidence that the superstructure was associated with the bismuth oxide layers in this material [25].

Because of the nontrivial materials problems presented by these materials, tunneling studies have tended to be controversial. Nonetheless, when the data from various studies are taken as a whole, the picture is much clearer. In Ref. 24 we undertook such a comprehensive look and concluded that there is considerable support for the large gaps we first reported in these materials, and that local tunneling probes seem more likely to see "clean" BCS-like tunneling characteristics.

We also used these films to study the critical current densities in these new superconductors. In the early days of the field, there was considerable concern over the very low critical current densities exhibited in bulk forms of the materials. Concurrently with work at IBM, we first showed that these superconductors could carry substantial critical currents, even at elevated temperatures [26]. This work provided a proof in principle that these materials could be used in high current applications. It remains true to this day that the best critical currents are obtained in these film samples. Part of the reason for this lies in their well-oriented microstructure—crucial in such anisotropic materials—but the origins of the high flux pinning, which must also be present, have not been identified.

In collaboration with the groups of John Clarke and Paul Richards at the University of California at Berkeley, we carried out studies of the noise properties of these films [27,28,29,30]. The work included flux noise relevant for SQUID applications and resistance fluctuations near Tc relevant for bolometer applications. This work clearly demonstrated the close connection between the noise and the perfection of the microstructure of the films. The noise performance distinctly improved as the degree of orientation of the grains improved, and was found to be low enough for applications when good *in situ* films were used. A proposal for a HTSC bolometer based on this work is described in Ref. 31. SQUID work at Berkeley is now underway based in part on the favorable prognosis provided by this work.

Other studies carried out on these films include measurements of the fluctuation conductivity above Tc and studies of the optical properties of these films. See complete list of publications.

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